
APPENDIX C

TECHNOLOGY DESCRIPTIONS

The technology options that the U.S. Department of Energy (DOE) has considered for the treatment of sodium-bonded spent nuclear fuel are described in this appendix. Each technology is described in the context of treating sodium-bonded spent nuclear fuel driver and/or blanket assemblies. A brief discussion of the technical maturity of each treatment technology is included at the end of each technology description. The technical maturity of the technologies range from mature technologies that have been previously demonstrated by DOE for spent nuclear fuel or in an industrial setting to immature technologies that have only been demonstrated on a laboratory scale or for which only a conceptual design has been developed.

C.1 ELECTROMETALLURGICAL TREATMENT

The electrometallurgical treatment process for sodium-bonded spent nuclear fuel was developed at Argonne National Laboratory for processing Experimental Breeder Reactor-II (EBR-II) driver and blanket spent nuclear fuel assemblies. The process has been demonstrated for the stainless steel-clad uranium alloy fuel used in that reactor. The electrometallurgical treatment process uses electrorefining, an industrial technology used to produce pure metals from impure metal feedstock (DOE 1996). Although most of the sodium-bonded spent nuclear fuel driver and blanket elements are composed of uranium metal alloys, there also are small quantities (about 0.1 metric tons of heavy metal) of sodium-bonded uranium oxide, uranium nitride, and uranium carbide fuel. The oxide fuel would be prepared for treatment using the electrometallurgical treatment process by reducing the uranium oxide to uranium metal with lithium metal dissolved in small batches of lithium chloride-potassium chloride molten salt solution. The resulting uranium-bearing solution would be added to the molten salt solution used in the electrometallurgical treatment process for other sodium-bonded fuel and blanket elements and processed with those materials. The carbide fuel would be prepared for electrometallurgical treatment by cleaning the fuel of sodium to the extent possible and then converting the fuel to uranium oxide with water or dilute acid. This oxide then would be converted to uranium metal by lithium metal in a molten salt solution and processed by the electrometallurgical treatment process with other sodium-bonded spent nuclear fuel and blankets. The nitride fuel also would be prepared for electrometallurgical treatment by converting it to uranium metal.

The description of electrometallurgical treatment in this environmental impact statement (EIS) is based on the assumption that the electrorefiner waste salts would be disposed of without salt recycling. This process differs from the original process described in the environmental assessment of the electrometallurgical treatment for the demonstration project (DOE 1996). In that assessment, the electrorefiner salts were to be treated in a series of zeolite columns. In these columns, the zeolite would absorb the fission products and transuranics from the salt and would release potassium as potassium chloride, which is one of the basic constituents of the electrorefiner salt. The bulk fluid handling system and zeolite columns were to be installed in the Hot Fuel Examination Facility argon cell. The potassium chloride salt and the recovered electrorefiner salts were to be reused in the electrorefiners. The fission products and transuranics that were absorbed in the zeolite then were to be removed from the columns in preparation for waste form production. Use of zeolite columns could potentially reduce the final ceramic waste volume.

Electrorefiner salt needs to be replaced if either the sodium concentration or the plutonium concentration limit is reached. The zeolite column would be a preferred option if the plutonium concentration in the salt became more limiting than the sodium concentration. In the latter case, which is the most likely scenario for the driver and Fermi-1 blanket spent nuclear fuel, the waste volume would be similar to the batch processing (i.e. without

salt recycling). During the demonstration project, due to lack of an available large-scale zeolite column, limited resources, and the fact that the batch processing produced acceptable waste forms and volumes, work focused primarily on batch processing. Additional research and development is needed to extend zeolite column use beyond the laboratory scale.

The individual steps in the electrometallurgical treatment process are described below. A diagram of the electrometallurgical process is shown in **Figure C-1**.

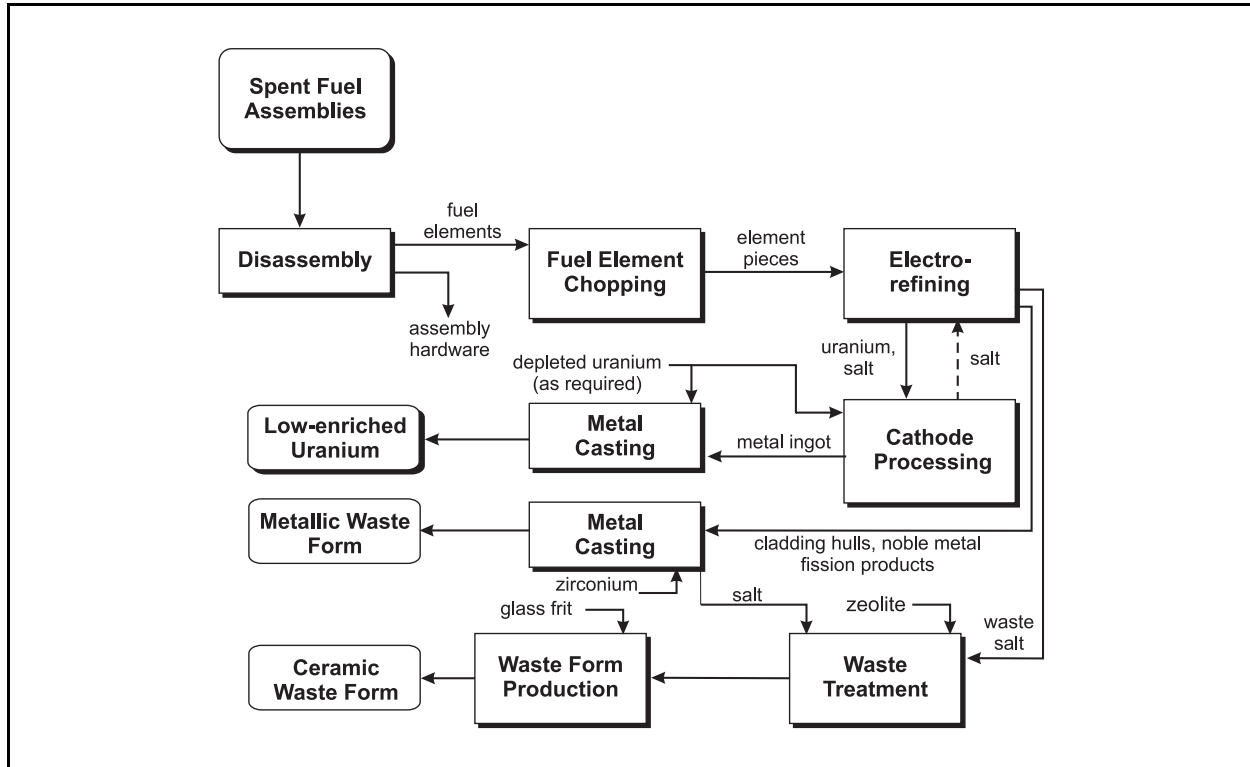


Figure C-1 Electrometallurgical Treatment Process Flow Diagram

Disassembly: Although the fuel and blanket assemblies mostly have been disassembled, some assemblies may need to be removed. The assembly hardware would be separated from the fuel elements that contain uranium and fission products by cutting the assemblies and physically separating the fuel elements. The fuel elements would be placed into a container for transfer to a hot cell containing an inert (argon) atmosphere for the remaining treatment steps. The assembly hardware would be stored at the Radioactive Scrap and Waste Facility at Argonne National Laboratory-West (ANL-W). This is a normal waste stream for ANL-W operations, and the separated hardware would be handled in accordance with normal site waste management practices.

Fuel Element Chopping: The sodium-bonded driver fuel or the blanket fuel elements would be placed in a machine for cutting into small pieces. The section of the element containing the fuel and sodium would be sheared into short segments. The section of the element containing the gas space (plenum) would be left intact. This section of the fuel pin cladding and the spacer wire would go into the metallic waste stream. The sheared fuel segments would be placed in perforated, stainless steel baskets to form an anode (positive electrode where oxidation would occur) for the electrorefiner.

During shearing of the hollow end (plenum) of the fuel pin, some fission product gases (primarily tritium and krypton) would be released to the argon cell atmosphere. These gases eventually would pass through high efficiency particulate air filters and be released up the emissions stack to the environment. All air emissions would be monitored and recorded.

Electrorefining: The electrorefiner is a machine in which the main electrometallurgical processes would occur. The electrorefiner vessel is made of steel. At its operating temperature of about 500 °C (930 °F), the vessel would contain a molten mixture of two salts, lithium chloride and potassium chloride. The electrorefiner also would have two or more electrodes: one or more anodes and one or more cathodes (negative electrodes where reduction would occur). Each anode would have baskets to hold the spent nuclear fuel pieces, and each cathode would consist of a bare steel rod where uranium metal would be collected.

The chopped fuel elements would be loaded into anode baskets and then lowered into the molten process salt. Upon application of an electric current between the anodes and cathodes, uranium, plutonium, and other transuranic elements, most of the fission products, and the sodium would be oxidized and dissolved into the salt. Uranium ions would be deposited at the cathode. Crystalline deposits of uranium would grow for 24 to 72 hours until almost all of the uranium in the anode baskets has been dissolved. The uranium-bearing cathodes would be raised into the gas space in the electrorefiner to allow some of the molten salt to drain away, although salt would adhere to each cathode. Each cathode then would be removed from the electrorefiner. The uranium deposit would be mechanically harvested and stored in the argon cell in a canister until it could be processed in the cathode processor.

The stainless steel cladding hulls and noble metal fission products would remain undissolved in the anode baskets. They would be removed from the electrorefiner and temporarily stored, prior to melting, into metallic waste-form ingots. The reactive fission products and transuranic elements would remain in the electrorefiner salt. The plutonium would be in a chloride compound in a liquid state and would be homogeneously mixed with other salts. The concentration of plutonium in the salt would be monitored through repeated sampling. The maximum plutonium concentration (about 8 weight percent) in the salt would not pose criticality safety concerns (Goff et al. 1999). In addition, abnormal localized concentrations of plutonium within the electrorefiner have been analyzed for a number of scenarios. These analyses have confirmed that an adequate margin of criticality safety would exist even under these conditions. The sodium would be in the form of sodium chloride (chemical form of table salt) as a part of the molten salt mixture.

The electrometallurgical process would use two electrorefining designs: Mark IV (for driver spent nuclear fuel) and Mark V (for blanket spent nuclear fuel). The Mark IV electrorefiner design would use a layer of cadmium to allow recovery of uranium that falls off the cathode during treatment. The Mark V design would use a collection basket instead of a cadmium layer.

Cathode Processing: The uranium deposits would be removed from the electrorefiner and treated to remove any adhering salt in the cathode processor, which is a furnace equipped with a vacuum system. The cathode product (along with depleted uranium to lower the enrichment of the resulting metallic ingot to less than 50 percent uranium-235, in the case of driver spent nuclear fuel) would be heated to about 1,200 °C (2,200 °F), melting both the uranium and the salt. Under vacuum conditions, the salt would distill away from the uranium and condense in a receiver crucible. The uranium would be melted in the cathode processor crucible and then solidified into an interim product ingot, which would be stored before final treatment in a casting furnace.

Uranium Metal Casting: The enriched uranium from driver spent nuclear fuel elements recovered in the electrorefiner would be melted together in a casting furnace with a separate stream of depleted uranium, electromagnetically stirred, and allowed to solidify. In this manner, enriched uranium from the treatment of driver spent nuclear fuel assemblies would be blended with depleted uranium in the casting furnace to form

low-enriched, metal ingots. The ingots then would be transferred to the Materials Storage Building within the Zero Power Physics Reactor complex, a controlled storage facility, until a decision is made by DOE regarding final disposition. Similarly, depleted uranium from treatment of blanket spent nuclear assemblies would be melted in a casting furnace and placed into storage until DOE makes a decision on final disposition.

| **Metallic Waste Form:** The metallic waste form is one of the two high-level radioactive waste forms generated
| from electrometallurgical treatment of sodium-bonded spent nuclear fuel. This waste form would consist of
| metallic ingots used to stabilize the stainless-steel cladding material, residual fuel matrix materials, and noble
| metal fission products. Actinides that remain in the cladding hulls after dissolution also would be present in
| the metallic waste form. These metals would be melted together in a separate casting furnace from the one used
| for uranium metal casting. Any salt remaining with the metals would be distilled away under vacuum at about
| 1,200 °C (2,200 °F). Upon heating to about 1,500 °C (2,730 °F), the metals would melt and form an alloy.
| A small amount of zirconium metal also would be added to improve performance properties and to produce a
| lower melting point alloy. After cooling, the metal would solidify into a metallic waste ingot. The typical
| composition of these ingots would be stainless steel, 15 weight percent zirconium, and about 1 weight percent
| noble metal fission products (Goff et al. 1999). These ingots would be packaged and stored in interim dry
| storage at the Radioactive Scrap and Waste Facility until shipment to a geologic repository in canisters for
| disposal.

| **Treatment of Electrefiner Waste:** At the end of a processing campaign, fission products and actinides would
| remain dissolved in the molten salt. The waste salt would be removed from the electrefiner and allowed to
| solidify. It then would be crushed and milled to obtain the desired particle size for ceramic waste form
| production. The liquid cadmium layer at the bottom of the electrefiner also would be removed periodically,
| filtered, and returned to the electrefiner. Filters from this bulk fluid handling system would become part of
| the metallic waste stream.

| **Ceramic Waste Form Production:** The ceramic waste form is the second waste form generated from
| electrometallurgical treatment of sodium-bonded spent nuclear fuel. The crushed and milled waste salt and
| dried zeolite would be added to a heated V-mixer. (Zeolites are crystalline aluminosilicates of group I (alkali)
| and group II (alkaline earth) elements. Their framework is a network of aluminum oxide and silicon oxide
| tetrahedra linked by the sharing of oxygen atoms. The networks of tetrahedra in the zeolite form cages in which
| molecules can be occluded.) The waste salt containing fission products and actinides would be absorbed into
| the crystal lattice of the zeolite, forming a dry particulate solid. Glass frit (sand-like glass) then would be mixed
| with the waste-bearing zeolite and placed in a special metal canister designed to be compressed to a desired and
| predictable shape. The mixture of material going into the process would be about 75 weight-percent waste-
| bearing zeolite and 25 weight-percent glass (Goff et al. 1999). This canister would be put into a type of furnace
| called a hot isostatic press, where it would be subjected to a temperature of 850 °C (1,560 °F) and a pressure
| of 1,057 kilograms per square centimeter (15,000 pounds per square inch). This would compress the canister
| and transform the material inside into a single cylinder of glass-bonded zeolite, which is referred to as the
| ceramic waste form. During compression, the zeolite would be converted to sodalite, a naturally occurring, salt-
| bearing material. Fission product chlorides largely would remain in the sodalite phase, while actinides (and
| most of the rare earth elements) would react with residual water in the zeolite to form oxide phases in the waste
| form. A conservative criticality assessment of the ceramic waste form indicated that the plutonium
| concentration in the waste form would pose no criticality safety concern (ANL 1999). These waste-form
| cylinders would be packaged and stored in interim dry storage at the Radioactive Scrap and Waste Facility until
| shipment to a geologic repository in canisters for disposal.

| **Technology Maturity:** The electrometallurgical treatment process is considered to be a mature technology.
| DOE demonstrated the electrometallurgical process for stainless steel-clad uranium alloy fuel used in the
| EBR-II reactor. Furthermore, it is an industrial technology used to produce pure metals from impure metal
| feedstock.

C.2 DECLAD AND CLEAN PROCESS

Cleaning (removing metallic sodium) and/or decladding are necessary steps in the treatment of sodium-bonded spent nuclear fuel using the plutonium-uranium extraction (PUREX) process at the Savannah River Site (SRS), the melt and dilute process at either SRS or ANL-W, and high-integrity can packaging. The fuel would need only to be cleaned of metallic sodium (i.e., it would not have to be declad) for melt and dilute processing at ANL-W and high-integrity can packaging. Decladding and sodium removal could be done using either a mechanical process (the melt, drain, evaporate, and calcine [MEDEC] process) or a laser declad and alcohol wash process. In the MEDEC process, the metallic sodium would be removed first; then, if necessary, the fuel would be declad. The MEDEC process has been performed for unirradiated blanket elements. The laser declad and clean process performs these functions in the reverse order—the fuel is declad using the laser and then the sodium is removed using an alcohol wash. Laser cutting is accompanied by partial volatilization of cladding, sodium, and materials dissolved or suspended in sodium, most notably cesium. This process was performed at the Rockwell International Hot Laboratory to declad and remove metallic sodium from approximately 7,000 EBR-II irradiated (low burnup) blanket spent nuclear fuel elements (Frazier and Campbell 1987). The process used a yttrium-aluminum-garnet (YAG) laser system. An automated fuel cutting sequence was developed, and the cladding was cut into strips. The cutting sequence included four circumferential cuts and three longitudinal cuts (at 120° circumferential segments) to allow mechanical removal of the cladding pieces. The bare fuel pins and the cladding pieces were washed in alcohol (ethanol) and water mixtures to remove the metallic sodium. The bare fuel pins were packaged and sent to SRS for processing. The contaminated alcohol mixture then was evaporated to reduce the volume, solidified with a grouting agent, and disposed of as low-level radioactive waste.

MEDEC Sodium Removal and Processing: This process would be performed at the Hot Fuel Examination Facility. Fuel elements would be brought into an argon-atmosphere hot cell where the ends of the elements would be cut off to expose the sodium within the cladding. The elements then would be cut into segments less than 61 centimeters (24 inches) in length. The fuel elements would be placed into a crucible and loaded into a closed induction furnace with an off-gas control system. The temperature in the furnace would be raised above the melting point of sodium (about 200°C [390°F]) and the molten sodium would be drained into a collection tank. With most of the sodium removed, the temperature would be raised to about 500°C (930°F) and a 10^{-4} torr vacuum would be applied to the chamber. This vacuum would volatilize the residual sodium, allowing the sodium vapor to be drawn away from the fuel. The vapor-phase sodium would be condensed in a trap and combined with the drained sodium in the collection tank pending further processing. Operating conditions necessary for complete sodium removal would be determined through testing. Verification of sodium removal would be obtained through analytical laboratory inspection and analysis. If the MEDEC process is applied for production, further verification would be performed. If necessary, the fuel pins would be mechanically pushed out of the stainless steel cladding after all the metallic sodium is eliminated.

Sodium recovered during the cleaning process would contain some fission products, most notably cesium-137. This cesium would be recovered by vacuum distillation of the sodium, taking advantage of the large difference in the boiling points of the two elements. The boiling point of cesium is 690°C (1,274°F), while the boiling point of sodium is 892°C (1,638°F). A vapor trap would be placed between the distillation column and pump to collect volatile species emitted from the condenser. The purified sodium would be processed by injection into a chamber where it would react rapidly with oxygen and water to form an aqueous sodium hydroxide. Carbon dioxide gas then would be bubbled through the hydroxide solution, converting the sodium hydroxide to a sodium carbonate. The aqueous sodium carbonate would be solidified with a binder and packaged for disposal as low-level radioactive waste. The cesium collected as distillate from the separation process would be added to the ceramic waste form described in Section C.1.

Laser Declad and Wash: As stated above, this process was performed at Rockwell International. The process would use a modified laser system for remote operations and a machine to hold and index the fuel elements

during cutting operations. The fuel elements would be brought into a hot cell. The fuel would be cut both circumferentially and longitudinally in a predefined cutting sequence. The fumes generated during the cutting process would be filtered and exhausted through an off-gas system. The fuel pins along with the cladding strips would be washed in an alcohol and water mixture to neutralize the metallic sodium and fission product (i.e., cesium) contamination. The fuel pins would be packaged and stored at ANL-W for further treatment, or sent to SRS for treatment. The alcohol and water solution would be evaporated partially, and the sodium/cesium alcoholates and hydroxides would be neutralized, then solidified in a grouting agent and disposed of as a low- or high-level radioactive waste, depending on the cesium content.

Compatibility with argon cell operation: The MEDEC procedure has been demonstrated using sealed vessels in an ANL-W facility. The laser process was demonstrated at Rockwell International. The laser process operation required personnel to enter the hot cell on a biweekly basis for laser maintenance and purging of the cell atmosphere to maintain a low oxygen level (less than 4 percent) and to vent alcohol-water vapors and hydrogen gas from the cell. Neither of these practices would be acceptable for argon cell operations. These concerns could be ameliorated by use of a hot cell with a different type of inert atmosphere, such as nitrogen, which could be purged and replaced with air. Nevertheless, personnel entry into the hot cell would still be restricted due to current radiation exposure controls and the higher level of fission products in the present inventory of sodium-bonded spent nuclear fuel. The use of alcohol and water in a multipurpose hot cell could raise a criticality concern if fissile materials were present in the cell. However, there would be no criticality issue with the blanket fuel itself, so a dedicated hot cell would eliminate this concern.

Finally, sodium collected during previous laser decladding operations was disposed of as low-level radioactive waste. The sodium collected from the processing of the fuel addressed by this EIS would be contaminated with cesium. If sufficient quantities of cesium were present in the sodium, this waste could not be treated as low-level radioactive waste. For the sodium to be managed as low-level radioactive waste, the sodium would have to be processed (as is done with the sodium removed from the fuel in the MEDEC process) to remove the cesium from the alcohol mixture using a currently undefined process. Because of the compatibility concerns associated with laser operation in the argon cell, the MEDEC process has been used to evaluate the various alternatives that require cleaning and/or decladding of the sodium-bonded spent nuclear fuel.

Technology Maturity: Argonne National Laboratory has used the MEDEC process to recover the uranium from 1,700 unirradiated sodium-bonded fuel rods. Laser decladding and cleaning were demonstrated on 17 metric tons of heavy metal (6,780 rods) of very low burnup EBR-II blanket fuel. Both processes are considered mature technologies.

C.3 PUREX PROCESS

The PUREX process is a counter-current solvent extraction method used to separate and purify uranium and plutonium from fission product-containing spent nuclear fuel and irradiated uranium targets. DOE has two facilities at the SRS, F-Canyon and H-Canyon, that use the PUREX process for the treatment of aluminum-clad fuel and targets. In this EIS, the PUREX process at F-Canyon is being considered for treating declad and cleaned EBR-II and Fermi-1 blanket spent nuclear fuel. The stainless steel cladding and sodium would be removed from these blanket spent nuclear fuel elements at ANL-W. The cleaned blanket spent nuclear fuel pins would be packaged in aluminum cans and shipped to SRS. The decladding and cleaning activities would be conducted in argon cells at ANL-W facilities. A diagram of the PUREX process is shown in **Figure C-2**.

Disassembly: The first step in the process would be similar to the disassembly process previously described in Section C.1. The assembly hardware would be stored at the Radioactive Scrap and Waste Facility at ANL-W and handled in accordance with normal site waste management practices.

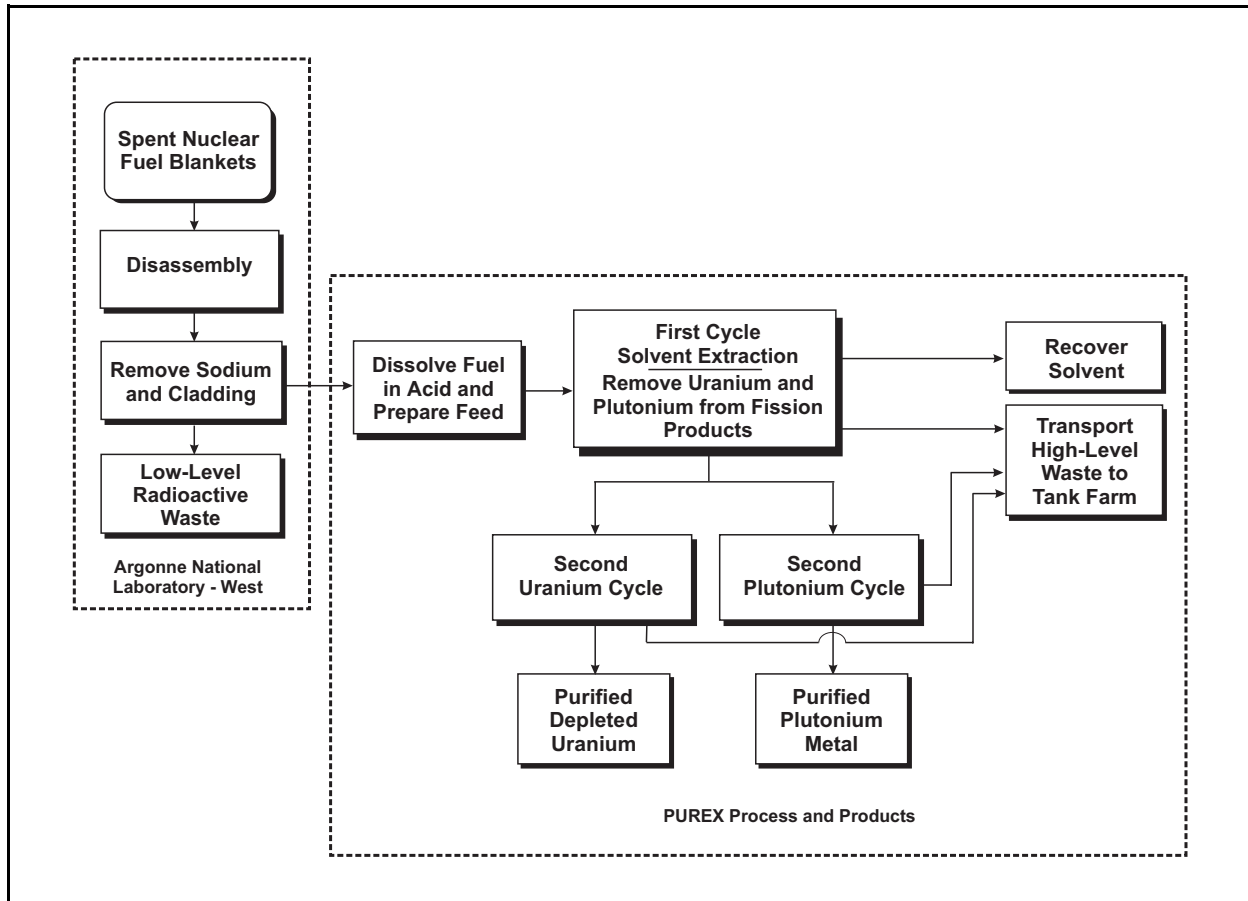


Figure C-2 PUREX Process Flow Diagram at SRS

Decladding and Sodium Removal: Blanket spent nuclear fuel elements would be cleaned and declad using the MEDEC process described in Section C.2. The uranium pins would be mechanically pushed out of the stainless steel cladding after all the metallic sodium has been eliminated. The bare uranium pins then would be packed into aluminum canisters in the Hot Fuel Examination Facility. The canisters, approximately 10 centimeters (4 inches) in diameter and 61 centimeters (24 inches) in length, would be backfilled with an inert gas and sealed. Each canister would contain about 60 kilograms (130 pounds) of depleted uranium fuel pins. The canisters would be placed in a NAC-LWT cask for shipment to SRS.

Receiving and Storage at SRS: The packages of blanket spent nuclear fuel pins from ANL-W would be received at the L-Reactor Disassembly Basin for storage until transfer to the F-Canyon for stabilization using the PUREX process.

PUREX Unit Operations: The EBR-II and Fermi-1 blanket spent nuclear fuel pins would be processed using the traditional PUREX process. This process consists of several major operations referred to as “unit operations,” which yield two products, uranium and plutonium (in solution form). The unit operations are dissolution, head end, first cycle, second uranium cycle, and second plutonium cycle. Unit operations that support the product recovery process are high-activity waste, low-activity waste, and solvent recovery.

Dissolution and Head End: The blanket fuel would be transferred to the canyon in casks and loaded into a large tank called a dissolver. Heated nitric acid in the tank would dissolve the blanket fuel, resulting in a solution containing depleted uranium, plutonium, and fission products. Gelatin would be added to the solution, if necessary, to precipitate fuel impurities. Then the solution would be transferred to a centrifuge

where impurities would be removed as waste. The clarified product solution from this process would be adjusted with nitric acid and water in preparation for the first cycle unit operation in the PUREX process. The waste stream generated from the process would be chemically neutralized and sent to the SRS high-level radioactive waste tanks pending further processing at the Defense Waste Processing Facility.

First-Cycle Operation: The first-cycle operation has two functions: (1) to remove fission products and other chemical impurities, and (2) to separate the solution into two product streams (i.e., uranium and plutonium) for further processing. This separation process occurs as the product solution passes through a series of equipment consisting of a centrifugal contactor and mixer-settler banks. Before the introduction of the product solution, flows of solvent and acid solution would be started through the equipment. After an equilibrium condition has been established, the product solution would be introduced. The chemical properties of the acid/solvent/product solutions in contact with each other would cause the fission products to separate from the uranium and plutonium. Later in the first cycle process, the plutonium would be separated from the uranium in a similar manner. The first cycle would produce four process streams: (1) a plutonium-containing solution (with some residual fission products), which would be sent to the second plutonium cycle; (2) a uranium-containing solution (with some residual fission products), which would be sent to the second uranium cycle; (3) a solvent stream, which would be sent to a solvent recovery cycle; and (4) an aqueous acid stream, which would contain most of the fission products and would be sent to the SRS high-level radioactive waste tanks pending further processing at the Defense Waste Processing Facility.

Second Uranium Cycle: In the second uranium cycle, the uranium-containing solution coming from the first cycle would be purified further in a manner similar to that described for the first cycle. The purified solution would be transferred to storage tanks. Eventually, the uranium would be converted to uranium oxide and stored in 208-liter (55-gallon) drums. The uranium oxide would be stored for future use. The solution containing the residual fission products would be sent to SRS high-level radioactive waste tanks pending further processing at the Defense Waste Processing Facility.

Second Plutonium Cycle: In the second plutonium cycle, the plutonium-containing solution coming from the first cycle would be further purified in a manner similar to that described for the first cycle. The purified solution would be converted to plutonium metal in the FB-Line. The plutonium would be disposed of in accordance with the Record of Decision (75 FR 1608), for the *Surplus Plutonium Disposition Final Environmental Impact Statement* (DOE 1999). The solution containing the residual fission products would be sent to the SRS high-level radioactive waste tanks pending further processing at the Defense Waste Processing Facility.

Other Unit Operations: The unit operations for high- and low-activity waste would reduce the volume of the aqueous streams containing fission products. The streams originate with primary separation process unit operations such as the first cycle. The fission products would be separated and sent to the high-level radioactive waste tanks. The volume reduction process would be accomplished using a series of evaporators in the canyons. The solvent recovery unit operation would recover and recycle the solvent used in the first cycle by removing impurities from the solvent. The purified solvent would be returned to the first cycle for reuse, and the impurities would be transferred to low-activity waste for processing (DOE 1994).

Technology Maturity: The PUREX process is considered to be a mature technology. It has been used throughout the world since 1954 to separate and purify uranium and plutonium from fission product-containing spent nuclear fuel and irradiated uranium targets.

C.4 HIGH-INTEGRITY CAN PACKAGING

The high-integrity can packaging option is being considered for EBR-II and Fermi-1 blanket spent nuclear fuel elements. The high-integrity can is made from Hastelloy Alloy C-22 metal alloy pipe having a 13.7-centimeter (5-inch) diameter, variable length, and a pipe wall thickness of 0.655 centimeters (0.258 inches) (Shaber 1998). Hastelloy Alloy C-22 is an alloy of nickel, chromium, and molybdenum that is highly corrosion-resistant due to its high chromium (22 percent) and molybdenum (13 percent) content. The high-integrity cans are designed for dry hot cell loading with a lid adaptable to wet loading and vacuum drying. The lid on each can has a threaded design to accommodate the partial loading of the spent nuclear fuel into the can at different times. The threaded lid prevents spillage of can contents during interim storage (DOE 1998). After packaging the fuel, the cans would be placed in standardized canisters of about 46 to 61 centimeters (18 to 24 inches) in diameter and up to 3 meters (118 inches) in length and would be codisposed with high-level radioactive waste in a repository. A diagram of the high-integrity can packaging is shown in **Figure C-3**.

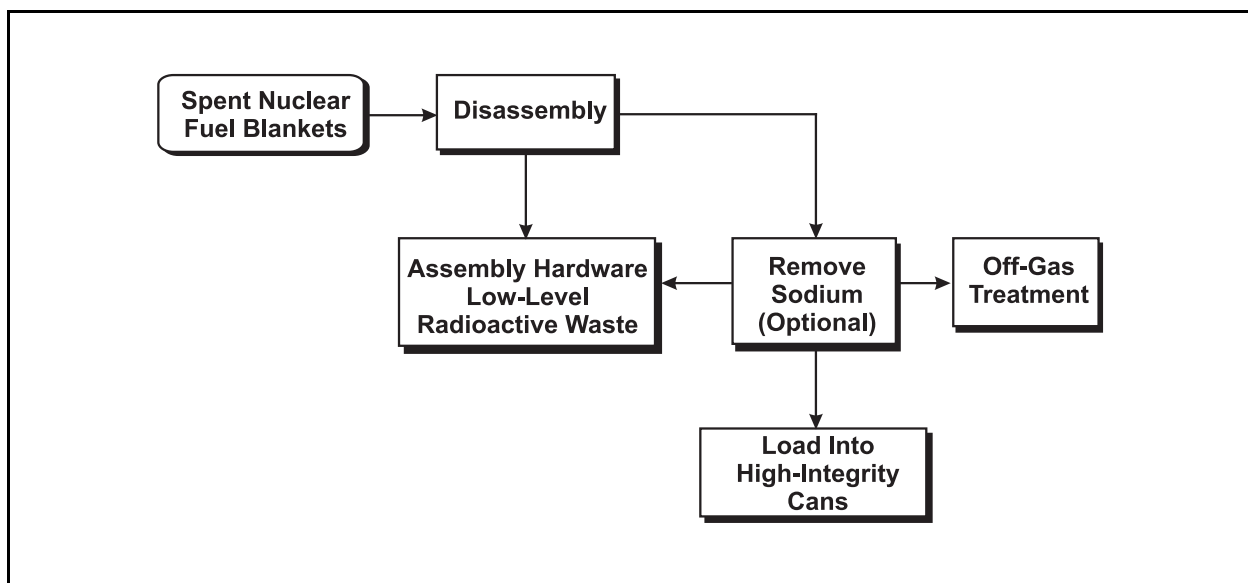


Figure C-3 High-Integrity Can Packaging Flow Diagram

Disassembly: Although the blanket assemblies have been mostly disassembled, there may be some assembly hardware that needs to be removed. The assembly hardware would be separated from the blanket fuel pins by cutting the assemblies and physically separating the fuel elements. The fuel elements would be placed into a container for transfer to an argon-atmosphere hot cell for the remaining process steps. The assembly hardware would be stored at the Radioactive Scrap and Waste Facility at ANL-W. This is a normal waste stream for ANL-W operations, and the separated hardware would be handled in accordance with normal site waste management practices.

Sodium Removal: If needed, the blanket spent nuclear fuel elements would be cleaned using the MEDEC process described in Section C.2.

Loading into High-Integrity Cans: The blanket spent nuclear fuel elements would be packaged in a standardized can fabricated from Hastelloy Alloy C-22, or possibly some other highly corrosion-resistant materials such as titanium Grade-12.

The high-integrity can would be placed in dry storage at an appropriate location. If transportation is required, the cans would be packaged into shipping casks. Prior to shipment to a geologic repository, the high-integrity

can containing spent nuclear fuel would be placed into a standardized canister, an overpack designed to provide additional containment within the waste package under repository conditions.

Direct Disposal of Sodium-Bonded Spent Nuclear Fuel: Direct disposal of sodium-bonded spent nuclear fuel is currently precluded by DOE policy concerning acceptance of Resource Conservation and Recovery Act-designated mixed waste (which contains both hazardous and radioactive waste). In the absence of such a policy, sodium-bonded spent nuclear fuel (driver and blanket) could be cleaned of surface sodium, packaged in high-integrity cans without removal of metallic sodium from the interior of the fuel elements, and directly disposed of in a geologic repository. The high-integrity cans would be placed into a standardized canister designed to promote containment under repository conditions.

Technology Maturity: Packaging materials in a high-integrity can is considered to be a mature technology. These cans would be made from highly corrosion-resistant materials and would be designed to provide exceptional protection from external environments.

C.5 MELT AND DILUTE PROCESS

The melt and dilute process is being considered for driver and blanket spent nuclear fuel elements. Three process options are being considered: (1) melting bare uranium blanket spent nuclear fuel pins with aluminum, (2) melting blanket spent nuclear fuel elements with cladding and additional stainless steel, and (3) using a modified melt and dilute process capable of handling the sodium in a volatilized form and processing chopped driver spent nuclear fuel elements that could not be completely cleaned of sodium. Processing activities would be conducted in the Hot Fuel Examination Facility at ANL-W or in Building 105-L at SRS. A diagram of the melt and dilute process flow for the first two options is shown in **Figure C-4**. A process flow diagram for the third option is shown in **Figure C-5**.

Disassembly of Blanket Spent Nuclear Fuel Elements at ANL-W: Although the blanket spent nuclear fuel assemblies mostly have been disassembled, there may be some assembly hardware that needs to be removed. The assembly hardware would be separated from the blanket spent nuclear fuel elements by cutting the assemblies and physically separating the fuel elements. The spent nuclear fuel elements would be placed into a container for transfer to an argon-atmosphere hot cell for the remaining process steps. The assembly hardware would be stored at the Radioactive Scrap and Waste Facility at ANL-W. This is a normal waste stream for ANL-W operations, and the separated hardware would be handled in accordance with normal site waste management practices.

| Sodium Removal and Processing at ANL-W: Blanket spent nuclear fuel elements would be cleaned using the MEDEC process described in Section C.2.

Decladding and Packaging Blanket Spent Nuclear Fuel Pins for Shipment to SRS: In the first melt and dilute processing option, blanket spent nuclear fuel pins that would be sent to SRS would be mechanically pushed out of the stainless steel cladding after all the sodium has been removed. These blanket spent nuclear fuel pins would be packed into aluminum cans in the Hot Fuel Examination Facility. The cans, approximately 10 centimeters (4 inches) in diameter and 61 centimeters (24 inches) in length, would be backfilled with an inert gas and sealed. Each can would contain about 60 kilograms (130 pounds) of depleted uranium spent nuclear fuel pins. The cans would be packaged and placed in a NAC-LWT cask for shipment to SRS.

| Receiving and Storage at SRS: The cleaned and declad blanket spent nuclear fuel cans from ANL-W would be received at the L-Reactor Disassembly Basin for storage until transfer to the processing facility in Building 105-L.

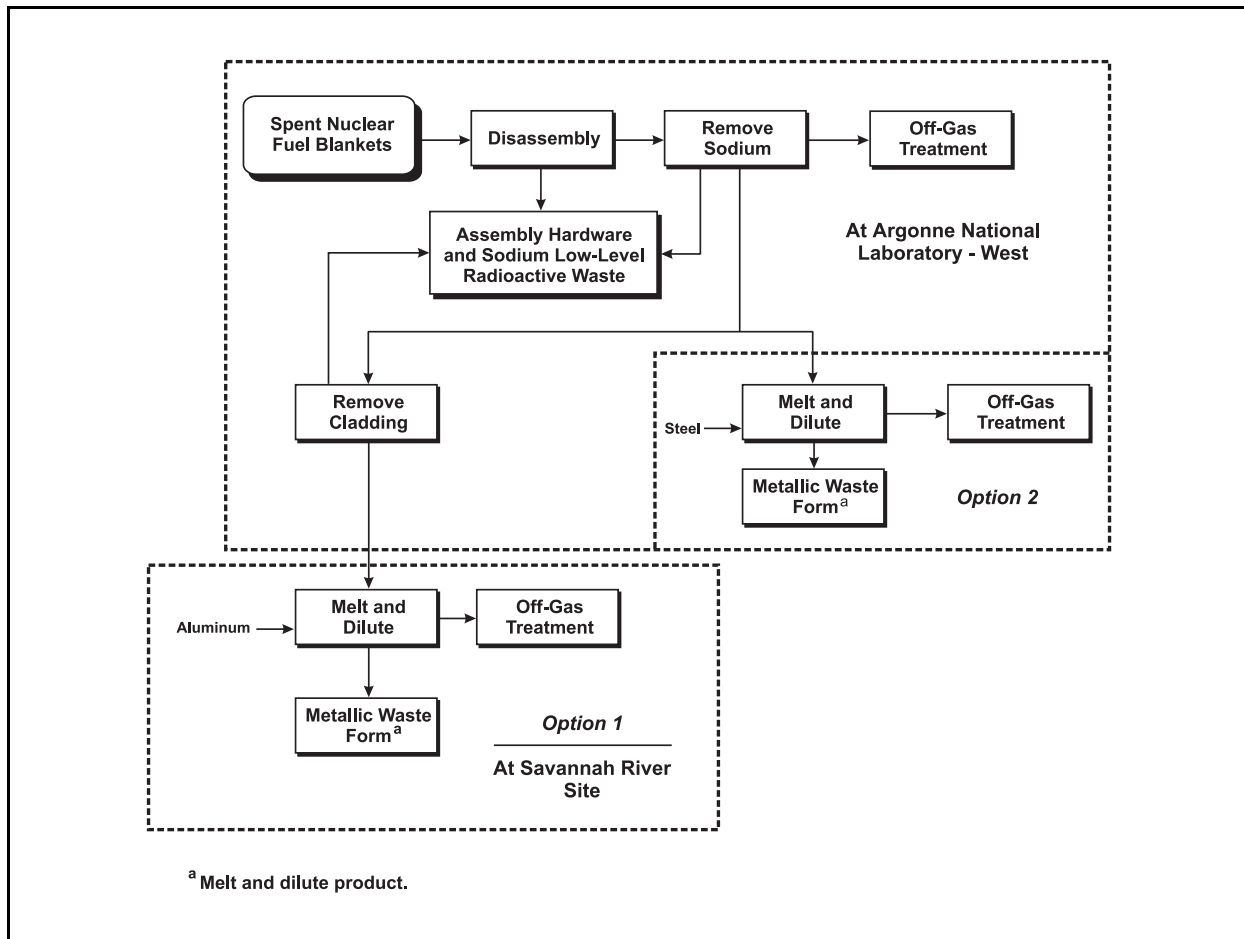


Figure C-4 Melt and Dilute Process Flow Diagram (Options 1 and 2)

Melt and Dilute Process for Blanket Fuel at SRS: Blanket spent nuclear fuel cans would be transferred to the treatment facility in Building 105-L for processing. The spent fuel cans would be loaded into an induction furnace where they would be heated to approximately 1,000 °C (1,830 °F). This temperature significantly exceeds the aluminum-uranium eutectic temperature required to initiate the melting, so it would proceed within a reasonable time. Sufficient aluminum would be added to make an aluminum-uranium alloy with a composition of about 70 percent aluminum and 30 percent uranium. The metal alloy would be cast into an ingot, sampled, and packaged into canisters. The canisters would be evacuated, filled with inert gas, sealed by welding, and transferred to storage pending disposition in a geologic repository. Volatile fission products would be captured by a series of filter banks before releasing the off-gas. The filters would be disposed of as low-level or high-level radioactive waste, as appropriate.

Melt and Dilute Process for Blanket Fuel at ANL-W: In the second melt and dilute processing option, blanket spent nuclear fuel elements recovered from the sodium removal process would be placed in an induction furnace crucible with additional radioactive waste steel. Sufficient steel would be added to make an alloy with a composition of about 50 percent each of uranium and steel. The furnace would be heated to approximately 1,400 °C (2,550 °F) to melt the uranium, after which the steel would be slowly dissolved into the uranium pool. The mixture would be electromagnetically stirred to a uniform composition. The metal alloy would be cast into an ingot, sampled, and packaged for interim storage at the Radioactive Scrap and Waste Facility. An off-gas system would capture the volatile and semi-volatile fission products for stabilization and processing into waste forms suitable for disposal. The filters would be disposed of as low-level or high-level radioactive waste, as appropriate.

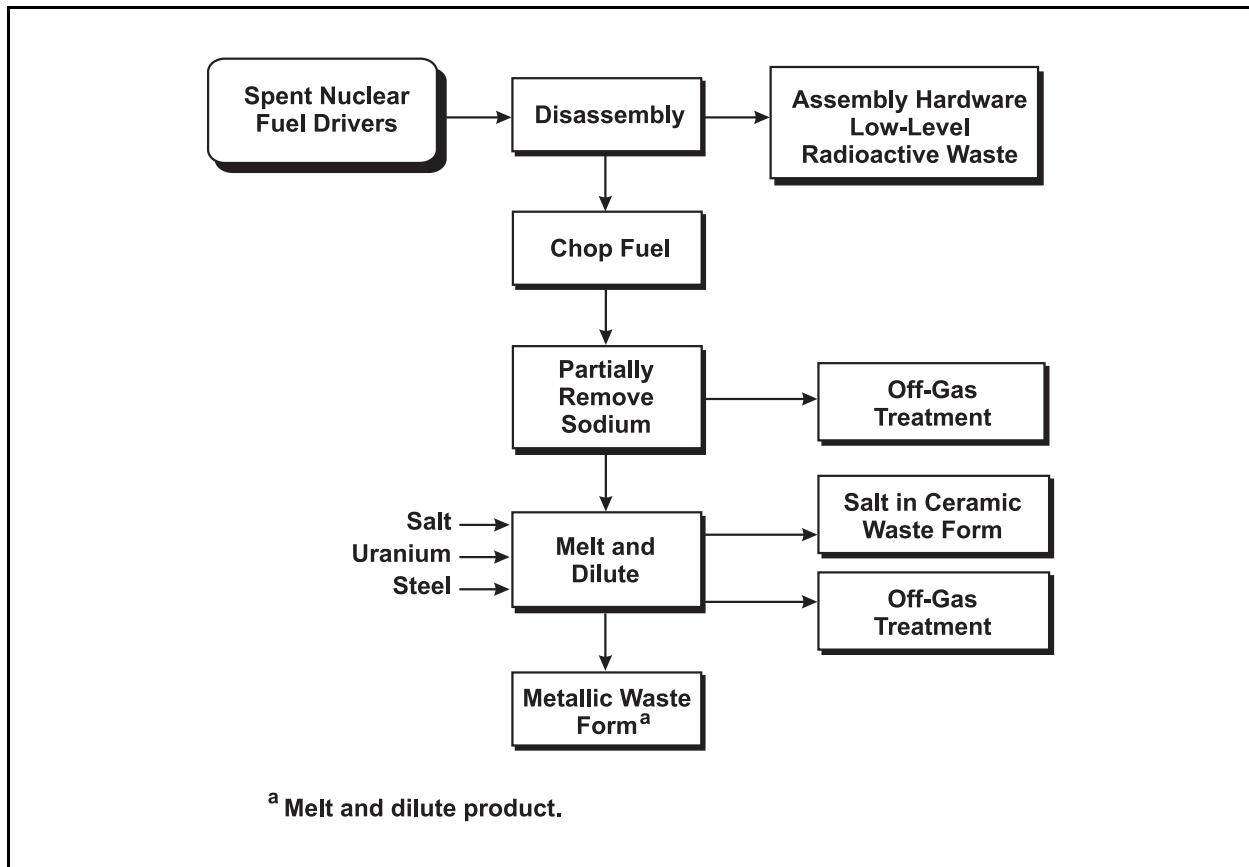


Figure C-5 Melt and Dilute Process Flow Diagram (Option 3)

Melt and Dilute Process for Driver Fuel at ANL-W: The third melt and dilute option would be for sodium-bonded driver spent nuclear fuel. Most of the metallic sodium in the driver spent nuclear fuel elements would be removed in a manner similar to the way sodium would be removed from the blanket spent nuclear fuel elements, i.e., the fuel would be cut into a few segments and heated to allow some sodium to drain away and then the fuel pieces would be heated under a vacuum to volatilize additional sodium. However, as explained below, not all the sodium could be removed by these processes. During the irradiation of the fuel in the reactor, after approximately a 1 to 3 percent burnup, the gap between the fuel pin and the cladding would be closed by the swelling of the fuel pin and interdiffusion between the cladding and the fuel pin. During the swelling process, the fuel pin would become porous and metallic sodium would enter the fuel. At discharge from the reactor, 15 to 20 percent of the fuel's pores would contain trapped sodium with dissolved fission products. The trapped sodium within the fuel pins and the areas of interdiffused fuel and cladding could not be removed.

Since not all the sodium could be removed from the driver spent nuclear fuel by the heating and vacuum process, a modified melt and dilute process would be needed. In this process, the driver fuel elements would be covered with a layer of low melting-temperature salt containing uranium chloride to oxidize the molten sodium. Depleted uranium would be added in a ratio of about 2.5 to 1 to reduce the enrichment to less than 20 percent uranium-235. Radioactive waste steel would be added in equal weight to the uranium to complete the mix. The furnace then would be heated to a temperature of about 1,400 °C (2,550 °F). The molten salt would capture sodium vapors escaping from the fuel elements as they melt, protecting the downstream components from the sodium. After volatilization of the sodium and reaction with the molten salt, a vacuum would be applied to the furnace to volatilize the salt, which would be condensed and partially reused. The

salt would be stabilized in the ceramic waste form described in Section C.1. The molten metal would be stirred to achieve a uniform composition, and then would be cast into an ingot, placed into a container, and stored. An off-gas system would capture the volatile and semi-volatile fission products for stabilization and processing into waste forms suitable for disposal. The filters would be disposed of as low-level or high-level radioactive waste, as appropriate.

Technical Maturity: The melt and dilute process was developed for treating aluminum-based spent nuclear fuel at SRS and is DOE's preferred technology for treating most (almost 97 percent by volume) of that type of spent nuclear fuel (DOE 2000). The melt and dilute process for stainless steel-clad spent nuclear fuel would require operating temperatures of approximately 1,400 °C (2,550 °F), compared with about 1,000 °C (1,830 °F) for aluminum-based spent nuclear fuel. Induction-heated melters that can achieve the higher temperatures required for stainless steel have been demonstrated at ANL-W. Technology development would be required to demonstrate capturing the quantities of sodium present in the driver spent nuclear fuel assemblies in a molten salt.

The melt and dilute process can be used for most of the driver spent nuclear fuel. However, there are small quantities (about 0.1 metric tons of heavy metal) of driver spent nuclear fuel that are composed of uranium oxide, uranium carbide or uranium/plutonium carbide, and uranium nitride, which have high melting points and cannot be treated using the melt dilute process.

C.6 DIRECT PLASMA ARC-VITREOUS CERAMIC TREATMENT PROCESS

The plasma arc treatment technology (DOE 2000) would use a plasma torch to melt and oxidize the spent nuclear fuel in conjunction with depleted uranium oxide and other ceramic-forming materials, as necessary. The fuel would be fed into the process with minimal sizing or pretreatment. The plasma arc would cut the fuel assemblies into small pieces and heat the fuel to temperatures at least as high as 1,600 °C (2,910 °F) to melt and oxidize it in a rotating furnace. Ceramic material would be added, as necessary, while the mixture was being homogenized by the torch. When melting and oxidation were complete, the rotating furnace would slow and the melt would fall into molds prepared to receive it. A diagram of the plasma arc treatment process flow is shown in **Figure C-6**.

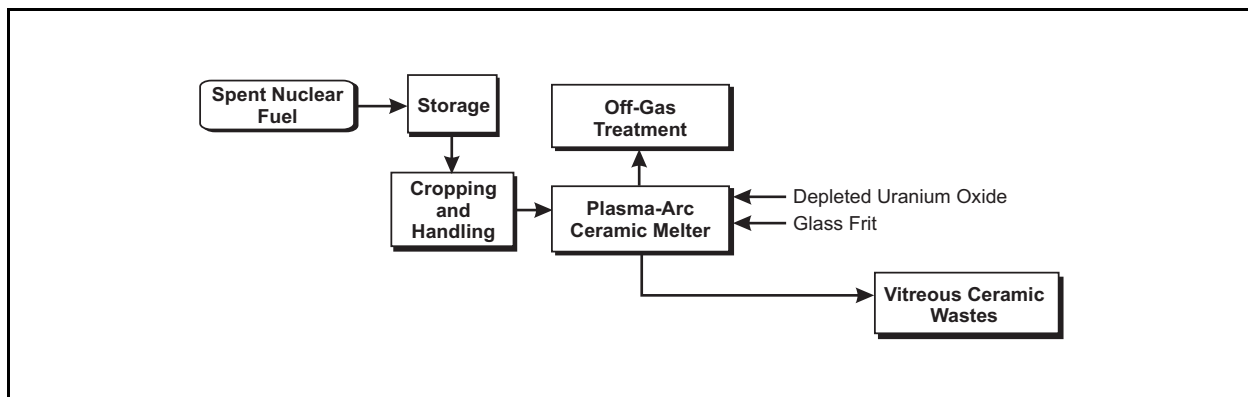


Figure C-6 Direct Plasma Arc-Vitreous Ceramic Treatment Process Flow Diagram

Metallic fuel such as EBR-II fuel would require the addition of some ceramic material. Depleted uranium could be added to the process in almost any form to reduce the uranium-235 enrichment. Criticality issues would be addressed by limiting the process to batch runs of preselected quantities of fissile material by the addition of the depleted uranium and by the addition of neutron poisons, if necessary.

As with all processes that dissolve or melt spent nuclear fuel, the plasma arc treatment would produce radioactive off-gases. These gases would be filtered and treated by appropriate means, with the filter and treatment media recycled into the plasma arc furnace for incorporation into the ceramic product.

Technology Maturity: The plasma arc process is a developmental technology that has not been demonstrated for stabilization of spent nuclear fuel.

C.7 GLASS MATERIAL OXIDATION AND DISSOLUTION SYSTEM

The Glass Material Oxidation and Dissolution System (GMODS) uses lead oxide to convert unprocessed spent nuclear fuel directly to borosilicate glass using a batch process. A diagram of the GMODS process flow is shown in **Figure C-7**.

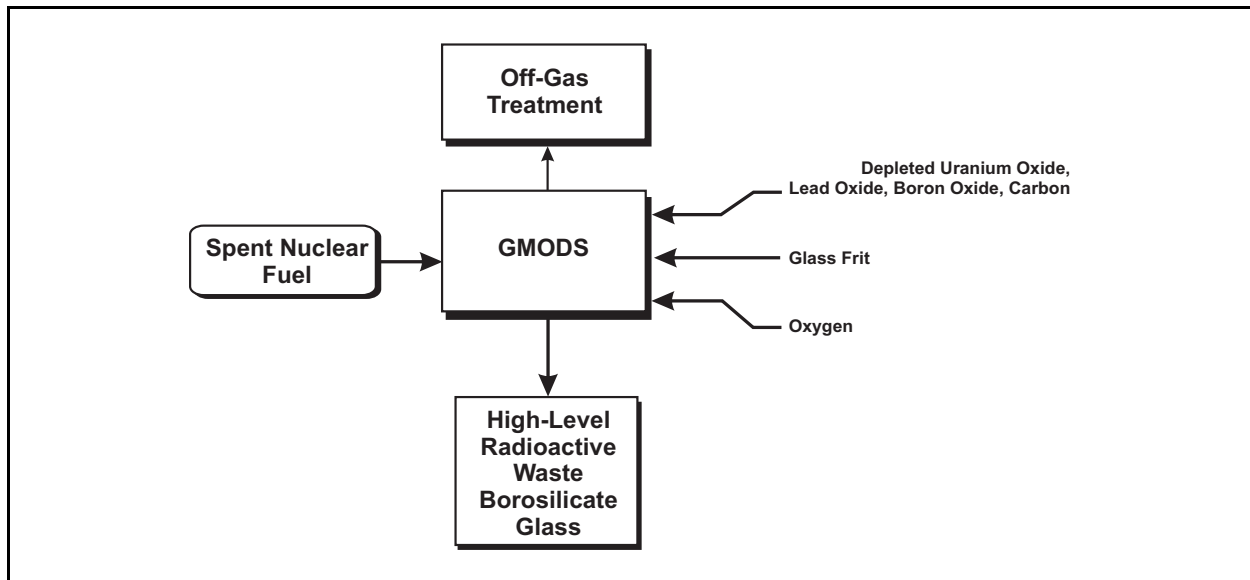


Figure C-7 GMODS Process Flow Diagram

Metal Oxidation: The principal piece of equipment for GMODS would be an induction-heated, cold-wall melter, which is used commercially to convert corrosive or high-melting metals to ultrapure materials. The melter, operating at 1,000 to 1,200 °C (1,830 to 2,200 °F), would be charged with a molten glass consisting of lead oxide and boron oxide. Oxides and amorphous components of the spent nuclear fuel would directly dissolve into the glass. Metals, which normally do not dissolve in glass, would be converted to oxides by the lead oxide. Boron oxide, a neutron poison, is a common agent for dissolving oxides into glass. Criticality concerns would be addressed by diluting the uranium-235 enrichment with depleted uranium and using boron oxide as a dissolving agent (DOE 2000).

On feeding the spent nuclear fuel into the melter, the uranium, plutonium, and other metals would be oxidized and dissolved in the molten glass. The oxidation of the metals would convert the lead oxide to metallic lead, which would sink to the bottom of the melter. Radioactive off-gases produced during this process would be filtered. The filters would be managed as high-level radioactive, low-level radioactive, or mixed waste, as appropriate.

Conversion of Lead to Lead Oxide: After decanting the glass, the melter would be recharged with boron oxide and, if necessary, lead oxide. Oxygen would be piped into the system to convert the metallic lead at

the bottom of the melter back to lead oxide. Therefore, lead would be an oxygen carrier that would not leave the system.

Glass Waste Form: The resulting glass mixture would not have qualities necessary for long-term durability, so silicon oxide (glass frit) would need to be added to increase the durability of the high-level radioactive waste borosilicate glass. The silicon oxide would not be part of the initial melter charge because its properties are not conducive to rapid oxidation-dissolution of spent nuclear fuel. Unreduced lead oxide could limit the durability of the glass and increase volume, so carbon would be added to the melt to reduce the excess lead oxide (DOE 2000).

Technology Maturity: The GMODS process was developed by DOE for stabilization of radioactive waste. At this time, it has only been tested in small-scale laboratory experiments.

C.8 CHLORIDE VOLATILITY PROCESS

Chloride volatility is an advanced treatment technology that was investigated at the Idaho National Engineering and Environmental Laboratory (NAS 1998). The process (1) uses the differences in the volatilities of chloride compounds to segregate major nonradiological constituents from spent nuclear fuel for the purpose of volume reduction, and (2) isolates the fissile material to produce a glass or ceramic waste form. A diagram of the chloride volatility process flow is shown in **Figure C–8**.

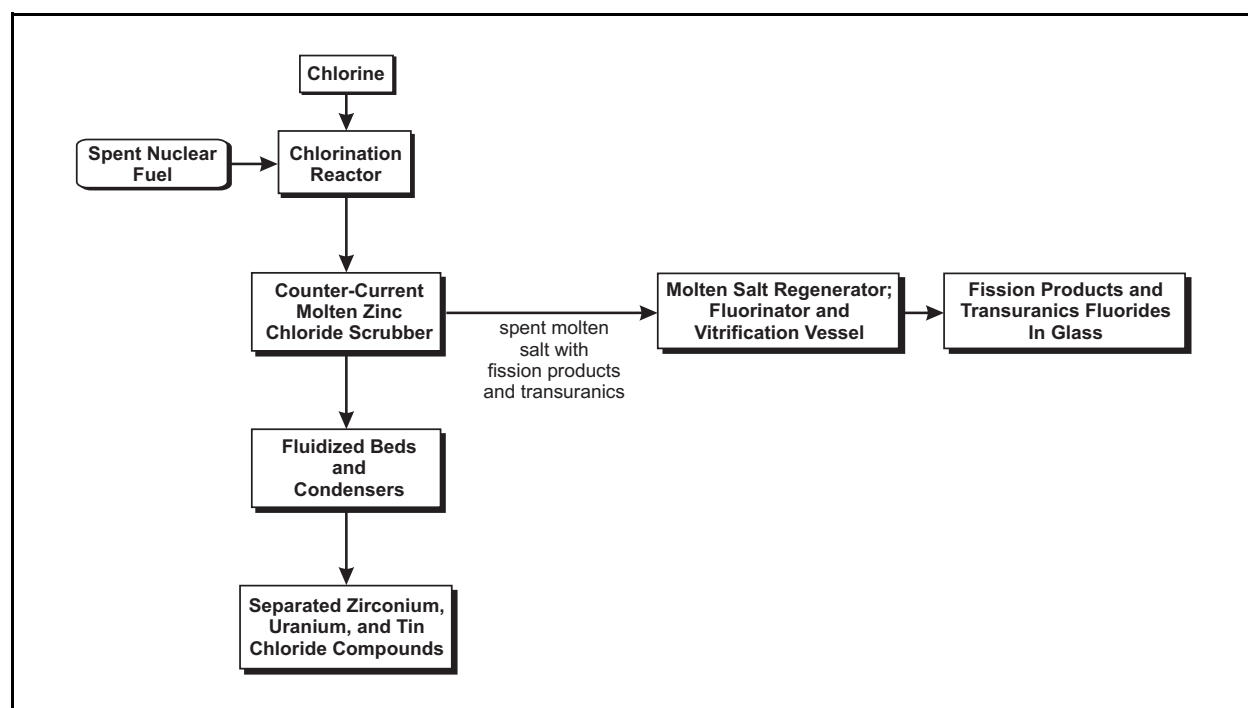


Figure C–8 Chloride Volatility Process Flow Diagram

The chloride volatility process would consist of four operations:

- (1) A high-temperature chlorination step that would operate at approximately 1,500 °C (2,730 °F) and would convert fuel and cladding materials to gaseous chloride compounds
- (2) A molten zinc chloride bed that would remove the transuranic chlorides and most of the fission products and would operate at approximately 400 °C (750 °F)

- (3) A series of fluidized beds and condensers that would operate at successively lower temperatures to condense zirconium tetrachloride, uranium hexachloride, and stannous tetrachloride
- (4) A zinc chloride regeneration/recycle process

The transuranic and fission product chlorides would be converted to either fluorides or oxides for final disposal. Argon carrier gas and unreacted chlorine gas would be recycled, the chlorine content adjusted, and the stream split and passed through the unit operations in a continuous closed loop. Periodic shutdowns of the coupled unit operations would occur for batch removal of fission product xenon and krypton gases from the carrier gas (such as by cryogenic distillation), batch transfer of the molten salt to the molten salt regenerator, and batch removal of nonradioactive constituents and uranium from the condensers.

The small quantity of fission-product/transuranic-product high-level radioactive waste would be converted into a waste form for repository disposal. The conversion steps to a glass or glass-ceramic form could involve fluorination and melting with glass frit additives, or conversion to oxides by heating at about 1,000 °C (1,830 °F) with boric acid.

In the chlorination step, the rate of reaction would be controlled by the feed rate of chlorine, and the temperature would be controlled by appropriate blending of argon gas with chlorine. An oxygen scavenger, such as carbon monoxide, would be added as needed to prevent formation of oxychlorides when oxides are present. A carbon dioxide absorption bed in the off-gas system would collect the carbon dioxide that would be formed. Zinc chloride would be used for the scrubber medium because its low melting point and favorable vapor pressure would permit its use to scrub the chlorinator off-gas at a low temperature, while its volatility at 725 °C (1,337 °F) would allow evaporative separation from the radioactive waste chlorides for subsequent recycle.

Theoretical chloride volatilities have been used to postulate the equipment sizing and operating parameters. Because of the lack of any experimental basis, significant concerns exist about the distribution of chloride compounds for multivalent elements such as uranium and plutonium. These concerns, in turn, lead to potential uncertainties in separation capabilities and overall flowsheet performance. The use of halides, either fluorides or chlorides, for the transuranic and fission product elements raises questions about the use of a glass or vitrified waste form. A proposal to use boric acid at about 1,000 °C (1,830 °F) allays some of those concerns (LITCO 1996).

Technology Maturity: The chloride volatility process has not progressed beyond the conceptual design stage. No laboratory experiments have been conducted.

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